U-PB DETRITAL ZIRCON GEOCHRONOLOGY OF THE LOWER DANUBE AND ITS TRIBUTARIES; IMPLICATIONS FOR THE GEOLOGY OF THE CARPATHIANS

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Key Points:

∗ A detrital zircon U-Pb study of modern sands from the lower Danube and its tributaries documents the main magmatic events that led to the continental crustal formation of the nearby Carpathians;

∗ The great majority of basement was formed in latest Proterozoic – Ordovician island arcs, a finding that is consistent with previous studies;

∗ An unexpected and prominent Carboniferous magmatic peak in the detrital record has no known source in the nearby Carpathians.

ABSTRACT

We performed a detrital zircon (DZ) U-Pb geochronologic survey of the lower parts of the Danube River approaching its Danube Delta- Black Sea sink, and a few large tributaries (Tisza, Jiu, Olt and Siret) originating in the nearby Carpathian Mountains. Samples are modern sediments. DZ age spectra reflect the geology and specifically the crustal age formation of the source area, which in this case is primarily the Romanian Carpathians and their foreland with contributions from the Balkan Mountains to the south of Danube and the East European Craton.

The zircon cargo of these rivers suggests a source area that formed during the latest Proterozoic and mostly into the Cambrian and Ordovician as island arcs and backarc basins in a Peri-Gondwanan subduction setting (~600 -440 Ma). The Inner Carpathian units are dominated by a U-Pb DZ peak in the Ordovician (460-470 Ma) and little inheritance from the nearby continental masses, whereas the Outer Carpathian units and the foreland has two main peaks, one Ediacaran (570-610 Ma) and one in the earliest Permian (290-300 Ma), corresponding to granitic rocks known regionally. A prominent igneous Variscan peak (320-350 Ma) in the Danube's and tributaries DZ zircon record is difficult to explain and points out to either an extra Carpathian source or major unknown gaps in our understanding of Carpathian geology. Younger peaks corresponding to arc magmatism during the Alpine period make up as much as about 10% of the DZ archive, consistent with the magnitude and surface exposure of Mesozoic and Cenozoic arcs.

KEYWORDS: Danube, Carpathians, detrital zircon, U-Pb geochronology, continental crust

1. INTRODUCTION

Although it is well established that the South and East Carpathians as well as the Apuseni and the Balkan mountains (comprising a Z-shaped double orocline of the easternmost part of the Carpathians and Balkans) were assembled during the Alpine orogeny a significant component of pre-Jurassic basement (Schmid et al., 2008, Matenco et al., 2010, 2017) records an older history that, in most places, is poorly known and sometimes controversial (Balintoni et al., 2014). A major obstacle is that more than 70% of the orogen is densely vegetated and thus poorly exposed. Some progress has been made over the past decade helped by modern geochronology data (see Balintoni et al., 2014 for a review of basement geochronology and the geological background below). This has led to most of the older interpretations regarding the origin and evolution of the Carpathians basement (Krautner, 1994, for a review) being revised or abandoned.

The relatively few zircon U-Pb geochronological studies of basement (pre Jurassic igneous and metamorphic) rocks (Balintoni et al., 2009, 2010, 2011, 2014, Balintoni and Balica, 2016) from each of the major geologic domains or their syn-tectonic cover rocks, (Stoica et al., 2016) have shown that the majority of basement rocks in the Romanian Carpathians and their foreland regions are Ediacaran to early Paleozoic island arcs. Confirmation and dating of Variscan magmatic and metamorphic rocks has also helped place the Romanian Carpathians within the regional geologic framework of nearby European basement terrains (von Raumer et al., 2013). But, despite these advances large areas, such as the Fagaras Mountains of the South Carpathians (the highest mountain range in the Carpathians), have not been visited by recent studies. As a consequence fundamental questions remain, such as whether there are Precambrian basement rocks to the Cambro-Ordovician arcs and if there was a succession of magmatic events associated with the Variscan collision. Future advances require new geologic and geochronological data as demonstrated for example by a recent study of a ductile shear zone in the South Carpathians basement (Ducea et al., 2016). Results in that study showed that terrane assembly took place during the latest Permian, much later in the evolution of the Paleotethys than previous models allowed and this changed understanding of the timing of metamorphism and terrane assembly of the South Carpathians. It is within this context that we conducted a DZ U-Pb study of modern river sediments from the Danube and its tributaries; our approach is a reverse engineering attempt at filling geochronologic/tectonic gaps in the scarcely known history of the regional basement through the lens of the sedimentary record of modern rivers. By comparing the known incomplete geologic record of the basement in the Carpathians with the limited but spatially significant collection of zircon ages from the most important rivers draining the Carpathian mountains (Radoane et al.,2003) and the lower Danube itself, we aim to detect what is missing from the regional geologic knowledge and where to target future localized studies of the basement.

Detrital zircon U-Pb geochronology is routinely used to investigate continental regions (Cawood et al, 2012; Gehrels, 2014). The ability to measure large numbers of zircons by in-situ mass spectrometry, mostly by laser ablation ICP-MS (Gehrels et al., 2008) has turned detrital zircon chronology into one of the most widely used quantitative provenance tools. Most studies aim to identify source area(s) of a sedimentary package by comparing zircon age distributions with bedrock ages from potential source areas (e.g. Barbeau et al., 2005; Thomas, 2011, Robinson et al., 2012; Gehrels and Pecha, 2014). Source regions are often distinguishable because each plausible source area has a specific geologic/tectonic history that includes different times, durations and fluxes of zircon producing magmatism and to a lesser extent, metamorphism. The goal of this study is the opposite in that we want to expand regional geochronological datasets by using modern river sediments to capture the zircon U-Pb age structure of rocks in river catchment areas. Here, we focus on the Danube and its tributaries that drain the easternmost segment of the Carpathian Mountains in Romania (Matenco et al., 2016). We found an unexpected abundance of Carboniferous (Variscan) zircons, a relatively young provenance age of the East Carpathian foreland, as well as some unexpected Eocene ages that among other data help to clarify existing hypotheses. Results also help guide future regional work.

2. GEOLOGIC BACKGROUND AND ZIRCON AGES

The Romanian Carpathians comprise a series of Alpine units each comprising several individual thrust sheets, stacked up during compressional tectonics. The main units are shown in the simplified map (Fig. 1 after Matenco et al., 2010, and previous work cited therein). From top to bottom, the major units are: (a) Tisza, which makes up the northern part of the Apuseni Mountains and parts of the Transylvanian Basin (Ciulavu and Bertotti, 1994), (b) east Vardar, a sequence of several thrusts that includes some primitive island arc rocks and pseudo-ophiolites of Jurassic age, (c) the Supragetic, making up the northern, western and eastern parts of the south Carpathians, (d) the Getic in the South Carpathians and equivalent Bucovinic thrust sheets in the East Carpathians (the Supragetic and Getic are sometimes collectively referred to as Dacia), (e) the Ceahlau-Severin thrust sheets represented in the South Carpathians by a narrow belt of serpentinites and attenuated flysch and in the East Carpathians by a larger flysch belt, (f) the Danubian, the lowest trust sheet package in the South Carpathians and (g) the thin skinned thrusts of the East Carpathians, which override the foreland to the east. In detail, these are complicated structures and have numerous alternate names and interpretations in the literature. In this study we follow the scheme of Matenco et al. (2010), which at the large scale is not fundamentally different from earlier syntheses (Burchfiel, 1976, 1980).

The compressional assembly of these distinct blocks took place between mid- and late Cretaceous during at least two poorly dated distinct tectonic events (confusingly referred to as “Austrian” and “Laramide” orogenic phases in the Romanian literature, Sandulescu, 1984), followed by a later sequence of thrusting in the East and South Carpathians, which started in the Miocene and extended into the Pliocene and Quaternary. The Apuseni Mountains, which contain internal evidence of Cretaceous thrusting, may have been translated from more southerly latitudes and has almost certainly been rotated clockwise during a Mid-Miocene (Balla et al., 1987; Patrascu et al., 1994; Dupont Nivet et al., 2005) episode of tectonic escape attributed to the Tisza bloc (Ratschbacher et al., 1993). Thus, the postulated position of the Apuseni Mountains on top of the other Carpathian units may be the result of a relatively young tectonic event. Clearly, the overall assembly of these thrust sheets is multiphase and their structural position today is complicated by translation along strike slip faults (Ratschbacher et al., 1993; Tischler et al., 2007; Ducea and Roban, 2016) and by the reactivation of some thrust faults as extensional structures (Schmid et al., 1998; Fügenschuh and Schmid, 2005).

For more detail on the tectonic elements and Alpine evolution of the Romanian Carpathians, which remain highly debated in the regional literature, we refer the reader to seminal papers by Schmid et al (2008), Matenco et al (2010; 2017), Csontos and Vörös (2004) and the earlier review by Burchfiel (1976), whose main points were made popular in the local literature by Sandulescu (1984). What is of importance to this paper is that the major units appear to contain thinned continental basement (igneous and metamorphic rocks of pre-Mesozoic age) and are separated by some relatively narrow basins in which sedimentation was marine (East Vardar, Ceahlau-Severin, and its later variant found in the East Carpathians, the Paratethys). None of these appear to have been part of the major Tethys ocean, whose main suture is located to the south of the Carpathians and the Balkans (Schmid et al., 2008). Instead they were basins possibly linked to the greater Tethys at times, formed on thinned continental crust and possibly containing small fragments of oceanic crust. This thinning presumably took place at the end of the Variscan orogeny, when a collisional belt collapsed in a Basin and Range-like fashion (Menard and Molnar, 1988), thus priming the Eastern European continental crust for the later development of the Tethys and related basins. Few of the Carpathian units described previously as ophiolites (Sandulescu, 1984, 1988) are geochemically or even geologically in the larger sense true ophiolites (Ianovici et al., 1976; Ionescu et al., 2009; Gallhofer et al., 2017). For example, the ones in the South Apuseni area, which were emphatically labeled as the “Main Tethysian Suture” by Sandulescu (1984), are actually rocks found in association with a predominantly calc-alkaline suite ranging from basalt to rhyolites (Gallhofer et al., 2017). In the broader sense these basins were back arc domains to the greater Tethys ocean, similar to basins of the Caucasus (Cowgill et al., 2016). Closure of these basins in the South Carpathians led to Alpine metamorphism (Ciulavu et al., 2008). Tisza, part of the east Vardar thrust sheet, the Supragetic, Getic, Bucovinic and the

Danubian units, all contain pre-Alpine continental basement, as does the foreland to the south and east. The thin skinned nappes of the East Carpathians do not have much exposed basement per se, but petrography of these units show clearly that they were sourced by rocks from the nearby foreland to the east.

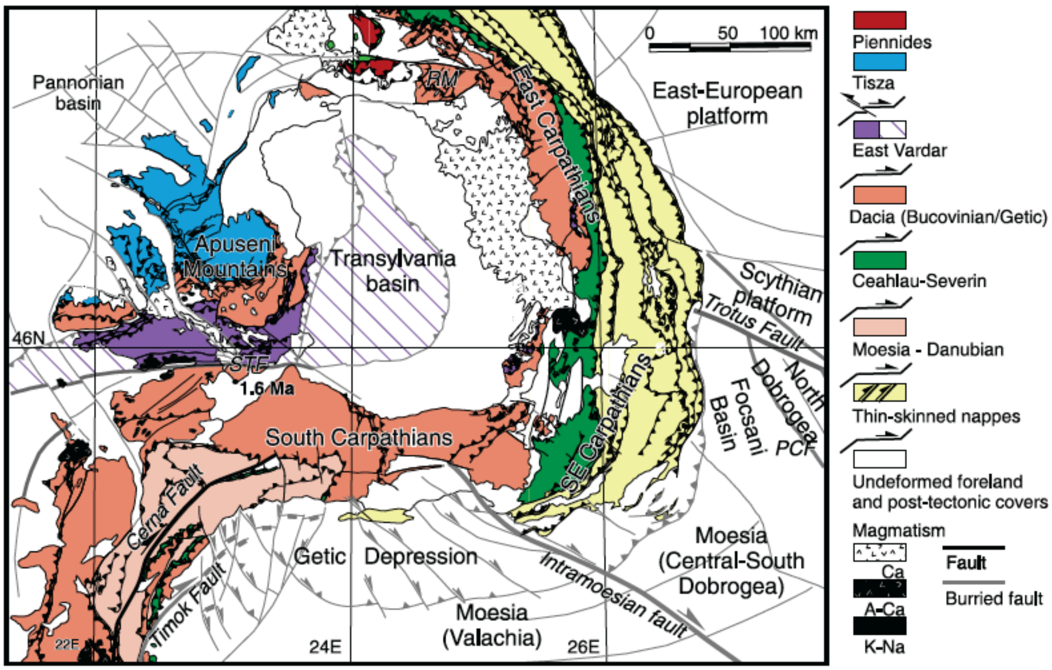

Figure 1. Modern configuration of the Carpathian orocline, with major geological and structural units, magmatic arcs (Ca=Neogene calc-alkaline, A-Ca= late Cretaceous calc-alkaline, and K-Na =Jurassic) and major faults. The map is compiled on the basis of the Geological Maps of Romania executed by the Geological Institute of Romania at various scales (1:1,000,000, 1:200,000, and 1:50,000) and subsequent work by the Free University of Amsterdam/University of Utrecht groups led by Prof. Liviu Matenco (e.g. Mațenco et al. 2010).

In the foreland there are several distinct blocks, some of which (Moesia, Scythia and the East European Platform) were viewed historically as platforms or even cratonic blocks based on the apparent lack of deformation of their cover rocks. In between them, lies North and South-Central Dobrogea, which are exposed in the province of Dobrogea (but continue unexposed under the Romanian plains) and clearly have a complicated Paleozoic and, in the case of North Dobrogea, even a Mesozoic history. The North Dobrogea does not represent a platform, whereas South-Central Dobrogea, which continues under east Moesia is considered by some as platformal. As more industry data (drilling and seismic) become available from the foreland regions of the Carpathians, it is clearer that none of these areas acted as rigidly as previously thought during the Alpine orogeny (Krezsek et al., 2017). In addition, they have a complex (and at the moment poorly resolved) Paleozoic history including magmatism as young as Carboniferous (e.g. in Western Moesia, Paraschiv, 1979).

While Moesia, Dobrogea, Scythia and even the western-most reaches of the East European platform close to the Carpathians remain highly debated in the geologic literature and poorly known due to lack of exposure, one aspect relevant to this study has become clear: none of these areas are legitimate cratonic areas free of deformation and magmatic activity for a sizable fraction of the Earth's history. Instead, as we will show below, they are dominated by Neoproterozoic U-Pb ages (560-610 Ma) and a distinctive array of less abundant 1-2 Ga zircons that are rather similar to the Danubian unit in the South Carpathians but very different from the higher Alpine structural units found towards the Carpathians interior.

Figure 2 is a compilation Kernel Density Estimate (KDE) type plot of igneous and (mostly) detrital zircons measured in various Carpathian and foreland units over the past decade. The great majority of these data were acquired at the Arizona Laserchron facility (and its precursors) at the University of Arizona and published by various groups cited in the figure. Recently, Gallhofer et al. (2015, 2017) have contributed to the U-Pb geochronology knowledge of the Jurassic and Cretaceous Carpathian magmatic arc rocks of South Apuseni and Banat, respectively. Older (pre-2008) zircon data from this segment of the Carpathians and its foreland are few: some TIMS work from the Edmonton laboratory in the early 2000s on a few Jurassic plutons of the east Vardar region (Pana et al., 2002), earlier TIMS work on Neoproterozoic Danubian granitic plutons (Liégeois et al., 1996) and limited 1960s-1980s geochronology performed in Soviet laboratories and published in local journals, without analytical details.

Zircon U-Pb ages from the interior of the Carpathians orocline (basement and cover units derived from them) are dominated by latest Precambrian (~600 Ma) to Cambro-Ordovician ages, in some areas continuing into the Silurian (Balintoni et al., 2009, 2010, 2011, 2013, 2014) (Figure 2). The following structurally higher units share this history: Tisza, East Vardar, Supragetic, and Dacia (Getic and Bucovinic). We refer to these as the Inner Carpathian Units. These rocks represent the products of relatively long-lived peri-Gondwana island arcs over that time period (600 – 420 Ma) with is a distinct dominant peak at 466 ± 10 Ma representing the culmination of a high flux magmatic event (Stoica et al., 2016). Older zircons are few and they make up a small Grenville peak and few ages older than 2.0 Ga- they are thought to be derived from a landmass closest to the island arcs. A less pronounced peak at 330±20 Ma represents the Variscan orogen and is believed to record a period of high grade metamorphism (Dallmeyer et al., 1998; Dragusanu and Tanaka, 1999; Medaris et al., 2003), which marks a continent-continent collisional event. Variscan zircons are mostly metamorphic. The end of high-grade metamorphism is tentatively constrained by the extension-related tectonic emplacement of some mantle peridotite into the crust (Medaris et al., 2003) at 316 ± 4 Ma.

In contrast with the interior units described above, the Danubian of the South Carpathians, all three Dobrogea blocks, the sedimentary units of the thin skinned thrust sheets of the East Carpathians (presumably derived from eastern sources) and the limited data on Moesia's basement all show a different U-Pb signature (Figure 2). They are referred to as the Outer Carpathian Units below and are found structurally in lower positions in the Alpine stack and toward or within the foreland. They have a late Variscan (285-300 Ma) signature, which in the exposed Danubian is represented by post-collisional often S-type granitoids commonly found elsewhere in the Peri Gondwanan basement of Europe (Stampfli and Borel, 2002; Stampfli et al., 2011; von Raumer et al., 2013) but which is not present in the inner units of the Romanian Carpathians (where the Variscan peak is at 320-350 Ma and is predominantly metamorphic). The dominant age peak of the Outer Units is Neoproterozoic one, 580±30 Ma with progressively fewer ages toward ~ 800 Ma. In the exposed Danubian, Neoproterozoic plutons are intruded into a metamorphosed sequence of 800 Ma island arc rocks (Liégeois et al., 1996); presumably a similar scenario applies to lesser exposed units of the foreland. Cambro-Silurian peaks (with a maximum at 460 Ma) which are common in the Inner Units do not exist here. The other distinctive feature of the outer units is the relatively abundant Proterozoic peaks at 1.2 Ga, 1.5 Ga, 1.75 Ga, 2.0 Ga and the late Archean peak at 2.7 Ga (Figure 2). While there is no evidence for basement of that age to have existed in the Danubian and probably the other outer units, they may have been located near to such a source (and the exact continental fragment representing that source remains debated, see Balintoni et al., 2014, Balintoni and Balica, 2016) whereas the inner units were not.

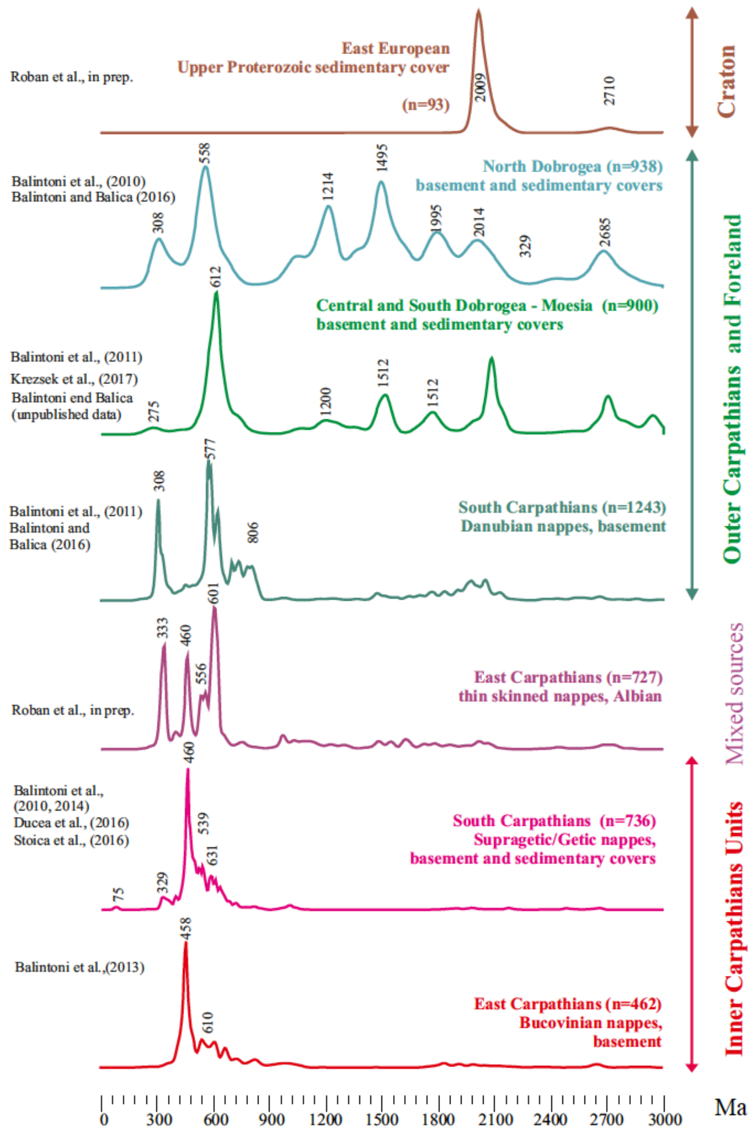

Figure 2. KDE (Kernel Density Estimate) age probability diagrams of Carpathians and foreland zircons (compiled from various published sources). The pink line representing sedimentary cover (thin skinned nappes) of Albian rocks combine zircons derived from Inner and Outer Carpathian units. The Outer Carpathians include the foreland domains to the east and south of the Carpathians

Overall, despite cursory knowledge of the geologic history of the Romanian Carpathian basement, all basement units above the Ceahlau-Severin suture (the Inner Carpathian Units) have a distinctive U-Pb zircon age distribution from the one found below it, including the foreland (the Outer Carpathians Units).

Alpine magmatism is relatively scarce; the Romanian Carpathians are covered by about 12% volcanic and intrusive rocks of post Permian rocks. A few early extension-related rocks in Dobrogea and the East Carpathians are known to be of Triassic age; the Ditrau alkaline massif of the East Carpathians (Ar-Ar age of 230 Ma, Dallmeyer et al., 1997) with a diameter of about 20 km is sizable, but unlikely to be a major source of detrital zircons.. The East Vardar region comprises latest Jurassic ophiolites and island arc rocks ranging in composition from basalt to rhyolites and syn- to late- granitoid plutons (Ianovici et al., 1976, Pana et al., 2002). These rocks occupy a sizable portion of the southern Apuseni Mountains but are probably zircon-poor due to their relatively mafic average composition. A belt of intermediate calc-alkaline rocks extending into the South Carpathians to the Apuseni Mountains, sometimes referred to as banatites (Berza et al., 1998), formed in response to the Sava ocean subduction to the south over a short period between 75 and 82 Ma in the Romanian segment (Zimmerman et al., 2008; Gallhofer et al., 2015). Although well studied for their numerous ore deposits and rich mineral diversity associated with them, outcrops cover only a small area. Mid Miocene volcanic and hypabyssal intermediate to silicic intrusives are found in the southern Apuseni Mountains (Seghedi et al., 2004, 2011; Rosu et al., 2004) mainly along an extensional lineament, but they too occupy a small fraction of the overall Carpathian region map area. Finally, volcanic and some hypabyssal intrusions of Miocene (~15 Ma) to Quaternary are found immediately to the west of the East Carpathians; they represent a sizable volcanic arc associated with the closure of the Paratethys ocean; the arc ranges in composition from basalt to rhyolite (Seghedi et al., 2011), and is on average an andesite. Given the catchment areas of the river sands sampled for this study (only the Tisza draws somewhat more heavily from rivers that cross cut this arc), Miocene or younger detrital zircons are unlikely to be common. Overall, these various plutonic and volcanic rocks are expected to be present in the river sands but only in low numbers.

One of the critical assumptions in the interpretations below is that the lower Danube in Romania and Bulgaria was closed in recent times to sediment supply from west of the

Carpathian barrier (the Golden Gate gorge) (Matenco et al., 2016) and thus the great majority of zircons measured in this study are sourced from the modern catchment areas in the nearby mountain range. This assumption may break down in detail as the Dacian Basin, for example (the foreland of the South Carpathians), may have been connected in the past (e.g. Miocene) to other segments of Paratethys and as such, may have recycled zircons from most distant sources contained in basin sediments. If zircons did come from outside of the Carpathian orocline, this assumption states it is unlikely they would be abundant.

3. SAMPLES

Fine to medium grained sand samples were collected from the recent alluvial deposits of the active bars and banks along the Danube River and four of its main tributaries (Siret, Olt, Jiu and Tisza) (Figure 3 and Table 1). Samples on tributaries were collected as close as possible to their confluence with the Danube.

Zircon concentrates were prepared using standard heavy liquids and a Franz Isodynamic magnetic separator set < 0. 5 amps. Zircon rich fractions were then mounted in epoxy resin and polished to expose internal surfaces. At no point were zircons hand picked as this can introduce bias. Analyses were made on every zircon like grain intersected during scanning along transects across polished grain mounts using a New Wave 193 nm aperture-imaged, frequency-quintupled laser ablation system coupled to an Agilent 7700 quadrupole-based ICP-MS. A typical laser operating condition for zircon uses an energy density of ca 2.5 J/cm^2 and a repetition rate of 10 Hz. Repeated measurements of external zircon standard Plesovice (TIMS reference age 337.13±0.37 Ma; Slama et al., 2008) was used to correct for instrumental mass bias and depth-dependent inter-element fractionation of Pb, Th and U. Temora (Black et al., 2003) and 91500 (Wiedenbeck et al., 2004) zircon were used as secondary age standards. Ages were based on the $^{206}Pb/^{238}U$ ratio for grains < 1000 Ma and the $^{207}Pb/^{206}Pb$ ratio for older grains. Data were processed using GLITTER 4.4 data reduction software and grains with a complex growth history or disturbed isotopic ratios, with > +5/-15% discordance, were rejected.

Data tables are provided in the Supplementary Material.

Table 1 . Sampling locations along Danube and tributaries.

Sample location	Latitude	Longitude	Distance to mouth [km]
Danube samples			
Tulcea	45°13'15.99"N	28°42'46.94"E	115
Brăila	45°19'22.59"N	28° 0'8.84"E	164
Turnu	43°42'46.30"N	24°53'28.30"E	596
Tributary samples			
Siret	45°23'56.80"N	28° 0'41.90"E	155
Olt	43°44'50.97"N	24°46'35.68"E	604
Jiu	44°34'09.80''N	23°27'19.80''E	694
Tisa	46°08'48.76"N	20°03'52.47"E	1214

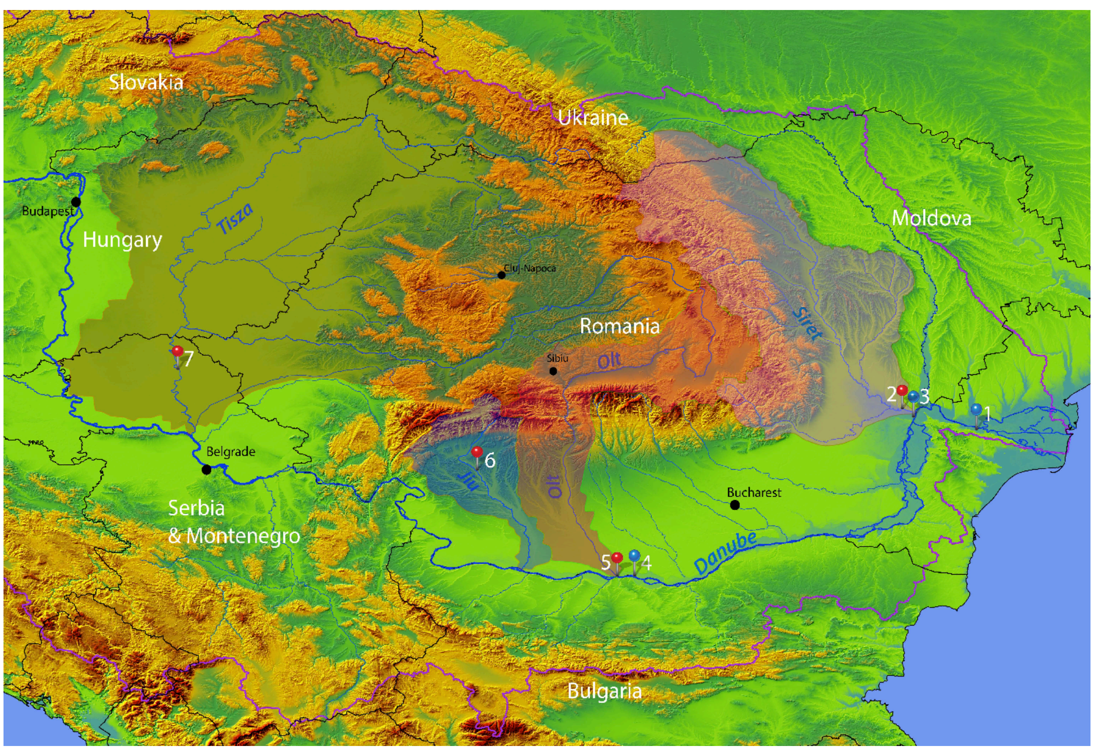

Figure 3. Sample locations along the Danube (blue symbols) and tributaries (red circles) from downstream to upstream: 1. Tulcea; 2. Siret River; 3. Braila; 4. Turnu; 5. Olt River; 6. Jiu River. 7. Tisza River. Catchment areas are show for each tributary sample. The map is color coded based on elevation- blue-green colors correspond to low elevations, whereas brown and red are higher elevations. The highest peaks are shown in the brightest of the red nuance.

4. RESULTS

Figure 4 shows KDE age distributions of Danube tributary samples and figure 5 shows KDE plots of Danube samples at different locations. The Kernel Density Estimate plots (Vermeesch, 2012) used an adaptive bandwidth. Below we present these results in detail and highlight the important age groups found in each sample. First, we discuss the results on the tributary samples followed by the presentation of the Danube samples. All samples are dominated by igneous zircons based on U/Th ratios (see supplementary data tables). U/Th ratios is excess of 10 are taken to reflect a metamorphic origin, whereas lower ratios (most commonly lower than 2-3) are typical of igneous zircons. Despite the fact that a dominant proportion of the Carpathian basement area is metamorphic, very few (<2% of the total analyzed population of zircons from the Danube and tributaries) zircons are metamorphic; they reflect the igneous crystallization of the protoliths. In previous studies (e.g. Balintoni et al., 2009, 2010), we observed that when zooming in at zircon rim scales (10 microns or less), metamorphic overgrowths are not uncommon among Carpathian basement zircons. In this detrital study, we measured only cores of zircon grains and did not focus on intragrain complexities.

4.1. Danube tributary samples

Tisza River

Tisza has a broad and complex provenance in that it mixes major tectonic units from the Bucovinic in the East Carpathians (mostly the Apuseni Mountains), with source areas of its tributary Mures, which draws from both the Getic-Supragetic and even the Danubian units. Therefore, the range of detrital zircon ages is expected to be diverse although dominated by zircons from the Inner Carpathian units. The Tisza age plot (Figure 4A) has a mixed age signal which is dominated by Inner Carpathians (Cambro-Ordovician 550-440 Ma with Variscan zircons 320-350 Ma) as expected, but it also shows significant input from the Danubian (290 Ma, which on Figure 4c can be seen as a secondary peak to larger 320 Ma and 600 Ma peaks). A few Precambrian zircons are also present. The most noteworthy feature, as is the case with the Olt River, is the predominance (50% of measured ages) of Variscan magmatic zircons, which is significantly different than one would predict based on existing data from any of the Inner Carpathians units. The presence of a few Jurassic zircons is attributed to the southern Apuseni island arc-MORB corridor whereas the late Cretaceous grains are part of the banatititc arc. Less expected are a few latest Permian to Triassic ages, which are difficult to correlate to known magmatic rocks in the Carpathians.

Jiu River

The river Jiu drains mostly Danubian rocks, with only a minor set of tributaries being sourced in the Getic unit. In this respect, the Jiu zircon population should be the simplest of all samples in this study and reflect the Danubian basement, which it does rather accurately (Figure 4C). The main age peak (575 Ma) corresponds to the dominant Danubian Neoproterozoic magmatic event found in the Danubian (Liégeois et al., 1996) as well as in other outer Carpathian units (Figure 2). Neoproterozoic ages decrease in number towards the 800 Ma age of the Dragsan Series, which is considered the oldest basement of the lower Danubian (Balintoni et al., 2011). Also present are some Cambro-Silurian ages from the Getic unit above which is drained by a few of the eastern tributaries of the Jiu. There is no evidence that such ages exist in the Danubian thrust sheets. There is also not a real Variscan *sensu stricto* peak at 330 Ma in the Danubian, apart from a couple of ages that may again be derived from the Getic unit. A second peak (290-300 Ma) corresponds to abundant late Variscan post-tectonic granitoids known from the Danubian. Older inherited age peaks at 1.0 Ga, 1.5Ga, 1.75 Ga, 1.95 Ga, 2.4 Ga and 2.7 Ga are also typical for Outer Carpathians units. The dominance of Neoproterozoic versus Permian ages in the detrital record is expected because the Jiu and its tributaries drain primarily the southern slopes of the South Carpathians, where the older ages prevail at outcrop. Rivers washing the northern slopes of the Danubian domain (e.g. Retezat Mountains) are more likely to be dominated by c. 300 Ma ages because of the high surface area occupied by a granitic batholith of that age.

Also present are a few late Permian and Triassic age (263 to 219 Ma) that do not fit with known Carpathian rocks. A few Mesozoic and Cenozoic ages are also present (see Supplementary Data Table as they do not properly show on Figure 4 due to the band width of the age spectrum) although these are attributable to the banatitic magmatism or Miocene tuffs that are known to be present in the foreland of the South Carpathians.

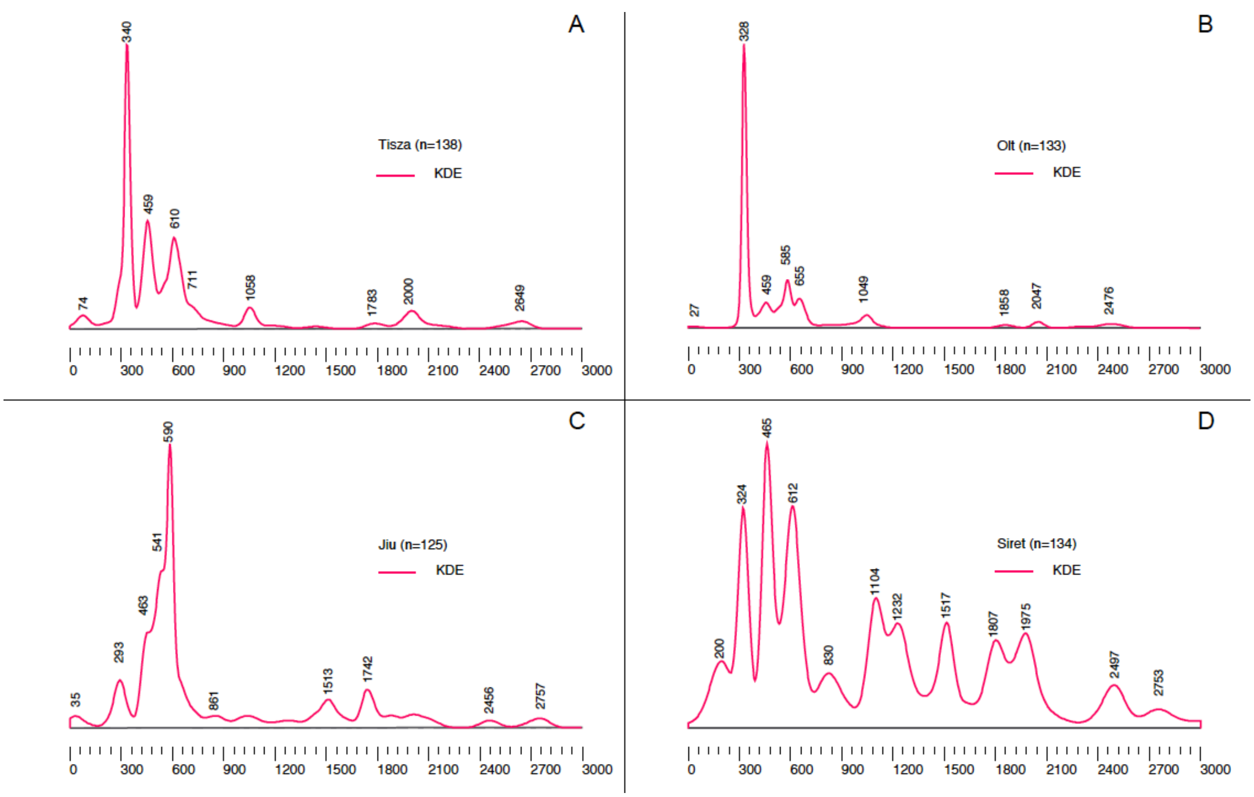

Figure 4. Danube tributaries river samples (A. Tisza, B. Olt, C. Jiu and D. Siret;) KDE (n = number of zircons ages). See text for further explanations.

Olt River

The Olt River drains mostly Getic and Supragetic units and since it originates in the South East Carpathians, it may contain some Bucovinic (Getic equivalent) in it. Some tributaries (Lotru or Cibin) may bring into the provenance mix material from the Danubian and southern Apuseni but it is subordinate compared to Getic-Supragetic sources. The greatest amount of sediment is driven from the glaciated Fagaras Mountains as well as the elevated areas of the central South Carpathians, all of which are Getic and Supragetic units and are overwhelmingly metamorphic (basement) rocks.

The Olt sample (Figure 4B) contains a large number of latest Precambrian to Cambro-Silurian ages typical for the Inner Carpathian units previously seen in the basement proper and in various sediments derived from it (Figure 2). However, the peak distribution is unlike the average of the Getic units. The surprise here is that more than Variscan (320-350 Ma) zircons dominate (≥ 50%) which is unlike any previous study of the Getic basement.

The Olt sample has the characteristically low abundance of older Precambrian peaks especially between 1.2 and 2 Ga (Balintoni et al., 2014), although there is a sizable Grenville peak (1-1.2 Ga) which is not common to Inner Carpathian units. A single late Eocene age is similar to Eocene zircons detected in some of the other samples but puzzling because such ages are unknown in any of the Carpathian units to the north of the Danube, or in the south in the Balkan Mountains. Eocene magmatism is prominent in the Rhodope Mountains south of the Balkans but this area does not drain into the Danube.

Siret River

The Siret River is the largest tributary of the Danube originating on the eastern slopes of the East Carpathians. Through its tributaries, the Siret's source areas include the Inner Carpathian Units as well as the outer Units and the East Carpathians foreland. The zircon KDE plot (135 zircons, Figure 4D) is complex and dominated by magmatic zircons, only two Precambrian grains show metamorphic Th/U ratios (>20). The Inner Carpathian Units diagnostic Cambro-

Ordovician-Silurian age pattern with a lesser group of Neoproterozoic ages is clearly distinguishable. Some zircon ages belonging to the Neoproterozoic peak may come from the Outer Carpathian Units. The range of Variscan ages suggest mixing of Getic metamorphic rocks (~ 330 Ma) with late Variscan S-type granitoids of the Danubian (290-300 Ma).

In addition, a more pronounced 1-2 Ga spectrum of ages is found in this sample compared to all others samples and relevant source areas (Figure 2). These ages make up more than half of the zircon age population. More than 5% are Archean (2.4 and 2.7 Ga), more than in other samples and is indicative of the Sarmatian craton as a source area. Taken together, these older than 1.0 Ga age peaks make up the largest grouping of pre Grenville zircons found in any basement or cover rock from the Romanian Carpathians and nearby foreland. Harder to explain are the presence of Mesozoic zircons. Eight zircons (making up 6% of the population) are Triassic, Jurassic or Cretaceous. Although there is a large Triassic alkaline massif (Dallmeyer et al., 1997) in the East Carpathians (Ditrau) the ages do not exactly match. Younger Mesozoic ages are also puzzling, and conceivably they may have been sourced from some of the Mid-Cretaceous flysch units of the Ceahlau unit (although there is no magmatic arc associated with the flysch units).

To summarize, the Siret river sand contains a diagnostic Carpathian signal but it also contains a significant number of Precambrian zircons and a group of Mesozoic ages that are not obviously tied to known source rocks within the river catchment area.

4.2. Danube samples

Danube at Turnu

The Danube at Turnu (103 zircons measured, Figure 5A) is a mix of sediment downstream of most inputs from most of the major South Carpathian rivers and this provides a good average of provenance prior to the arrival of rivers from the East Carpathians (e.g. Siret) and from more East European cratonal sources (Prut). In addition to being located downstream from the major Carpathian rivers Turnu is also found downstream of the largest Bulgarian rivers draining the Balkan Mountains. Variscan intrusions are abundant in the Balkans (Carrigan et al., 2005) but their age span, between 317- 297 Ma, is distinctively younger than our magmatic ages that fall between 350-320 Ma.

The Turnu sample appears a mixture between a Jiu-like (Danubian) KDE and an Olt-like (Getic-Supragetic) age distribution. Both Neoproterozoic (at around 850 Ma) and late Variscan granitoid ages are present although Arguably Danubian sources are slightly more important than Getic-Supragetic ages supported by distinct Mesoproterozoic peaks. The unexpected Variscan peak (igneous zircons of 320-360 Ma) found in the Olt River sample is also present and has the same magnitude relative to the Cambro-Silurian peaks of the Getic-Supragetic units. A few Cenozoic ages (45 Ma, 23 Ma, 6 Ma) are unlike any igneous activity known in the mountainous regions representing the source of the lower Danube at this location. It is possibly that some tuffs or loess derived from them, found in the foreland of the Carpathians and other lesser studied volcanic units in the Balkans to the south could prove to the sources of these zircons. At this point, however, these ages do not match the existing Cenozoic regional geologic record and are difficult to use forward.

Danube at Braila

This sample (118 zircons measured, Figure 5B) location was chosen to give an integrated Danube signal prior to the arrival of the last two rivers from the Moldovan foreland, some of which could carry a much more East European cratonal age signature (dominantly Archean see Figure 2) compared to the ones derived from the Carpathian and Balkan orogens. Otherwise the signal should not be much different from that at Turnu.

The bulk of of the detrital zircon age distribution is consistent with a mix of Inner and Outer Carpathians plus foreland sources (including the now ubiquitous magmatic Variscan signal from the Getic-Supragetic). It is clear that overall no one event dominates, neither the Neoproterozoic of the Danubian, nor the Cambro-Silurian arcs of the Getic/Supragetic, nor the Variscan ages of the Getic (possibly also from the Balkans in the south), or the post Variscan granitoids of the Danubian. In all, they contributed more or less similarly to the zircon budget and are consistent with erosion of the modern Carpathians. A few young outlier ages exist at Braila, some derived from the Neogene Volcanic field (<10 Ma), other representing either the Eocene ages seen in other samples or the well-established latest Cretaceous magmatism.

Danube at Tulcea

This sample (104 individual zircons measured, Figure 5C) represents the integrated Danube DZ signal downstream from the arrival of the main cratonal tributary (Prut) and just before entering the Danube Delta to drain into the Black Sea. The breakdown of ages is about 40% Inner Carpathian Units, 40 % Outer Carpathian Units, 10% Alpine ages (Jurassic East Vardar, late Cretaceous banatitic magmatism, Neogene magmatism including two unexplainable Oligocene ages), about 5% inferred to be from the nearby North Dobrogea terrain (based on the dominance of 250 Ma igneous ages there, Balintoni and Balica, 2016) and the remainder probably being derived from either East European craton or more likely peri-cratonal areas similar to the Outer Carpathians but dominated by 1-2 Ga ages. Only two of these zircons (dated at 420 and 322 Ma) have high Th/U rations and are therefore metamorphic in origin. The integrated Danube signal shows that the Inner and Outer Carpathians each contribute about half of the present-day zircon cargo despite the greater exposure of Inner Carpathian units in the mountainous regions. The integrated signal undoubtedly contains zircons that were derived from sedimentary sources such as the thin-skinned nappes of the East Carpathians that primarily came from Outer Carpathian Units. Overall the great majority of the Carpathians and foreland were formed in the latest Proterozoic to the middle of the Paleozoic (600-420 Ma) followed by the enigmatic Variscan (320-350 Ma) episode of magmatism, which still makes up about 17% of the integrated signal at Tulcea.

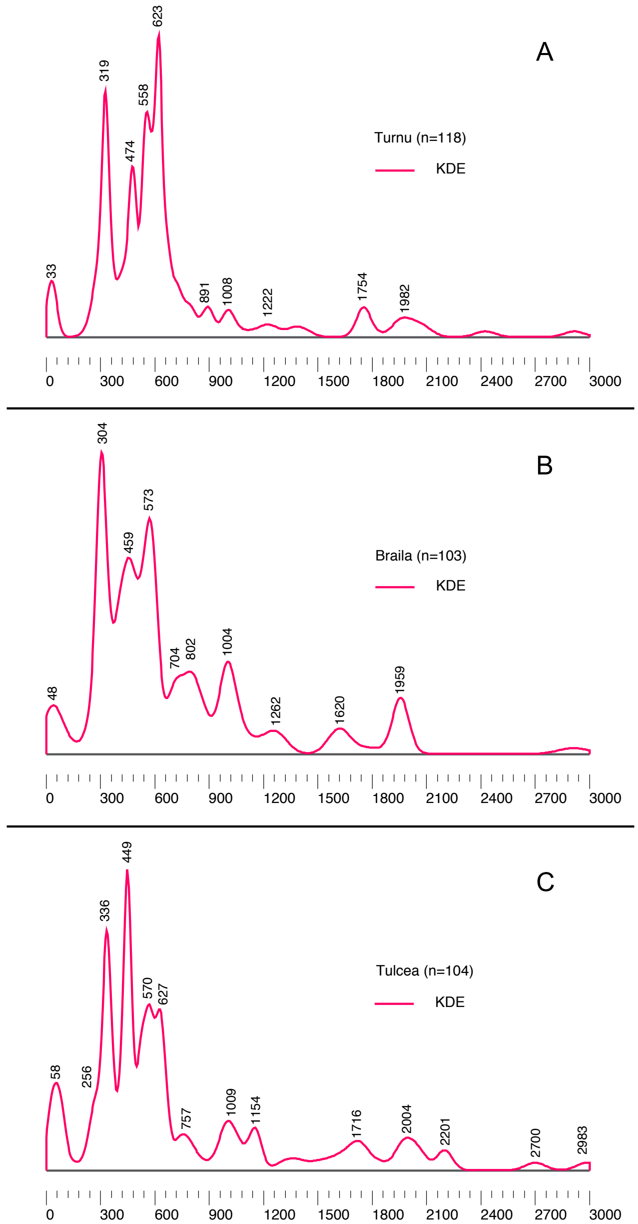

Figure 5. Danube River samples (A. Turnu, B. Braila, C. Tulcea) KDE (number of individual zircons shown in the diagram). See text for further explanations.

5. INTEPRETATIONS AND IMPLICATIONS

Comparison of previously published data from the Romanian Carpathians basement and results from this study allow us to make a crude estimate of the igneous and metamorphic events responsible for the making of this segment of continental crust (Figure 6). Below we detail our findings and uncertainties associated with them.

5.1. Nature of basement

The main results of this study confirm previous work that suggest all Carpathian units and the foreland formed during the Neoproterozoic and early Paleozoic in a series of island arcs and marginal basins formed in a peri-Gondwanan setting (Balintoni et al., 2014). This scenario applies to much of the pre Alpine basement of mobile Europe (Stampfli et al., 2011; von Raumer et al., 2013). Results also support the view that age structure and composition of the Inner Carpathian units are different from the Outer Carpathian units and foreland. The Inner units have little inheritance from earlier Precambrian zircons and have two stages of major crustal growth, one at 560 Ma and the other at 460 Ma. These are followed by well-known Variscan barrovian metamorphism and some poorly documented associated magmatism (320-350 Ma). The Danubian unit in the South Carpathians and the foreland (including Dobrogea), are better represented by inherited ages in the 1-2 Ga interval (Balintoni et al., 2012; Balintoni and Balica, 2013), and record a distinct magmatic age between 570-620 Ma, along with a lesser but important age peak at around 800 Ma (the oldest rocks of the Carpathians),. There is also a distinct episode of post collisional magmatism at 290-300 Ma, some post Variscan S-type granitoids known regionally in the Danubian but no obvious Variscan metamorphism (315-350 Ma) as seen elsewhere. The study detrital zircon age distributions of the main Danube tributaries match these established events and can therefore be considered representative of the regional geology.

Each of these two major domains (separated by Alpine basins, now closed as sutures) contributes about 37% of the total DZ signal of the Danube as it enters its delta. The remainder is made up of some Alpine magmatic ages, limited craton input from the Eastern European stable area to the north and an unexplained but sizable (17% of the total DZ budget) group of Variscan magmatic ages not known in the Romanian Carpathians or Variscan magmatism in the Balkan Mountains to the south (see below). These ages aside, the bulk of the Carpathian continental crust was formed in island and transitional arcs and other marginal (e.g. backarc) basins close to Gondwana, between about 600 and 420 Ma, and with a dominant age peak at around 460 Ma. The oldest arc is found in the Danubian unit and is a mafic island arc remnant of about 800 Ma.

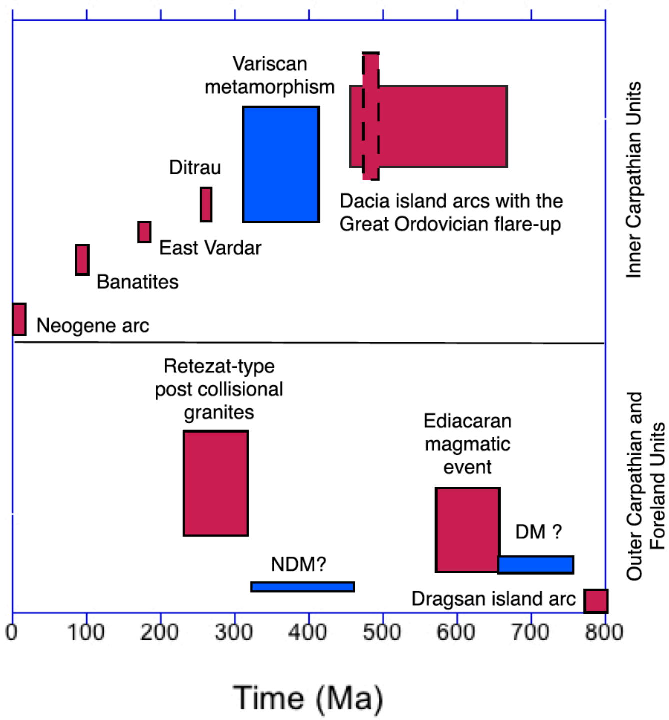

Figure 6. Major magmatic (red) and metamorphic (blue) events in the Romanian Carpathians and foreland, as evident from this and previous studies. The enigmatic Variscan magmatic event described in this paper is not shown here. DM – Dragsan metamorphism (age unresolved but prior to Ediacaran magmatic event). NDM – North Dobrogea metamorphism, also age unresolved. Vertical length of the boxes shown in this figure is proportional to the zircon abundance budget in the rivers studied here, whereas the extent of metamorphism is based on the surface exposure of these metamorphic rocks relative to magmatic rocks.

5.2. Ages of the East Carpathian foreland

It is impossible to quantify how much of the river signal is foreland-derived since almost all of the Carpathian foreland is covered by younger sediment and vegetation. The Dobrogea forebulge (Matenco et al., 2013) is located in the foreland to the Carpathian oroclinal bend and its zircon budget is somewhat similar to the Outer Carpathian units (Balintoni et al., 2014, Balintoni and Balica, 2016). Ultimately, we still know very little about the basement of Moesia and the Eastern European foreland east of the Carpathians and this study was not designed to add much to that. However, a noteworthy feature is that the Siret River, which drains the East Carpathians and a significant part of the East European foreland, has a DZ pattern similar to Outer Carpathian units and is not dominated by craton sources from the east. Rivers to the east, such as the Dniester in Ukraine and Moldova and certainly the Volga and Don in Russia, may provide a more accurate craton signal. Recognizing that the Siret river DZ signature probably mostly derives from the thin skinned nappes and not the foreland, we argue that the immediate foreland to the east Carpathians is not part of the craton. The Scythian and other poorly known mobile belts bordering the East European craton (Kuznetzov et al., 2014) must make up the majority of the basement of the Moldovan foreland, while legitimate cratonic Archean blocks are nowhere near.

5.3. Variscan ages

The Variscan (315-350 Ma) igneous (low Th/U) ages so prevalent in the analyzed samples, represent the most surprising finding of this study, based on the known regional geology. Limited numbers of metamorphic ages are to be expected but not magmatic ages. There are three potential explanations: (1) magmatism of Variscan age is more prevalent in parts of the Carpathians basement than previously recognized, (2) these grains were brought into the foreland basins (e.g. the Dacian basin, or Moesia) from a southerly origin (the Balkans) and recycled in the modern Danube and tributaries, and (3) these ages are extra-Carpathian, i.e. brought by the Danube or paleo-Danube from significantly upstream where Variscan magmatism is more widespread (Neubauer and Handler, 1999; von Raumer et al., 2013).

The first explanation is highly unlikely despite our limited U-Pb geochronologic knowledge of large areas of the South Carpathians, in particular the Fagaras Mountains. While the glaciated and elevated Fagaras Mountains may represent a major source of sediment in the Olt River, and only one sample (detrital, Balintoni et al., 2009) has ever been analyzed for U-Pb ages from the northern slopes of the range, it is clear that they are not made up of predominantly igneous rocks, but instead they are dominated by various metamorphic sequences (Pana and Erdmer, 1994). Garnet Sm-Nd geochronology data (Dragusanu and Tanaka, 1999) shows conclusively that metamorphism is Variscan (320-360 Ma). All areas from the Romanian Carpathians undergoing Variscan modifications are characterized by amphibolite grade metamorphism that ended at about 315 Ma (Medaris et al., 2003) with extensional collapse; neither the peak metamorphism nor the extension that followed it is associated with significant felsic plutonism. Many of the high grade rocks of Dacia contain some leucogranite and pegmatites (Hann, 1995), but they represent less than 1% of the exposed area (Horst Hann, personal communication, 2017), are Permian (255-280 Ma) in age and have distinctively large Th/U as high as 300 (our work in progress). It is thus unlikely that a yet to be identified Carpathian terrain can be the source of these zircons.

The second and third possibilities presuppose that Variscan zircons were transported into the peri Carpathian Paratethys basins at an earlier time either from the south (the Balkan Mountains) or west of the Romanian Carpathians (various locations in central Europe) and later incorporated as local sources into the lower Danube and tributaries (see Figures 3 and 6 in von Raumer et al., 2013). The Balkan mountains do contain, in contrast to the Carpathians, significant areas of Variscan magmatism (Carrigan et al., 2005), but their age is somewhat younger (317-297 Ma) than the Variscan peak identified in this study. We do not identify the Balkan ages with those in our data.

An exo- Carpathian source – to the west and beyond the Carpathian double bend – is plausible for the Danube itself if it can carry coarse material through the Iron Gates gorge, which is debatable. But for the tributary rivers, such as the glaring example of Olt, an Outer Carpathian source would presume that the Dacian basin fill (part of the Paratethys in recent times) contains relatively far traveled zircons from times when this basin was interconnected to others of the Paratethys (Pannonian, etc.) (Matenco et al., 2013). Those Variscan zircons were then transported further downstream by the Olt River from the Miocene-Pliocene sedimentary fill of the Dacian basin. Alternatively, in the so-called “spill and fill” model (Bartol, et al., 2012, Leever et al., 2010, 2011), a paleo-Danube that formed upstream in the Pannonian basin infilled parts of the Dacian basin and Variscan zircons are now eroded and carried by the Olt. The puzzling fact that the Jiu River, which also traverses the Moesian foreland (the Dacian basin) does not have such Variscan ages may be explained by a rapid infill of the western Dacian basin by local rivers (Fongngern et al., 2016). The “concurrent basin fill” model (Olariu et al., 2018) calls for Carpathian rivers infilling most of the Dacian Basin, which would have limited the import of zircons from beyond the Iron Gates .

The more than 50% Variscan ages found at the mouth of the Olt River, the dominance of the same peak in the Tisza sample and smaller but significant fractions found downstream along the Danube (with about 17% of the total zircon being Variscan at Tulcea, the terminus point of Danube before entering the Delta) remains unresolved and puzzling. Future studies should investigate this in more detail using detrital records, especially from along the Olt River and its archive immediately to the south of the South Carpathians and into the Moesian plain.

5.4.Younger ages

Clearly, only a small fraction of the DZ ages in the Danube’s archive is made of Alpine ages. About 10% of the mountainous terrain in the Carpathians consists of Mesozoic and younger magmatic rocks, and the Danube budget of zircons reflect roughly that (although the East Carpathians so called Neogene volcanic belt is located in an unusual position relative to the hydrographic network and is located far from the samples studied here). Of the Alpine magmatic rocks of the Carpathians and Balkans, the East Vardar island arc and its MORB-like basement are not significant zircon producers (because there are abundant gabbros and mafic rocks), the late Cretaceous Banatitic arc is poorly exposed in a couple of narrow belts that are unlikely to produce a large zircon cargo, and the Neogene volcanism in the East Carpathians and Apuseni Mountains are most volumetrically significant. Regardless, the Neogene arc only provides a minuscule number of zircons along the lower course of the Danube, as expected.

They are volumetrically overpowered by the main crustal-forming peri Gondwanan magmatic arcs and marginal basins of both the inner and outer Carpathians and its foreland.

Two minor but unusual groups of ages stand out among the Alpine ones and generate two additional problems in matching zircons sources to sinks : Triassic and Eocene ages. The only Triassic magmatic bodies known in the Romanian Carpathians and North Dobrogea, respectively, are the alkaline massif of Ditrau and a suite of small alkaline bodies in western north Dobrogea assumed to be of similar age. However, the existence of a few Permo-Triassic ages in almost all analyzed samples suggests that Triassic magmatism may be more prevalent in the Carpathians. We have some evidence that suturing of Paleozoic terranes in the South Carpathians continued into the Triassic (Ducea et al. 2016) and that the Getic pegmatites are of that age as well. Either other early extension plutons similar to Ditrau (but volumetrically smaller) exist in the Carpathian nappes or post collisional pegmatites provide this age range in the detrital record.

Eocene ages (40-30 Ma) are even more puzzling because there is no Cenozoic magmatism of that age in the Romanian Carpathian realm (Seghedi et al., 2011). An Eocene arc is well developed in the Rhodope mountains to the south, but the current hydrographic network or even known ancient ones (Matenco et al., 2013) do not link that source area to the Danube or its lower tributaries. Possibly earlier links between various basins of the Paratethys (Matenco et al., 2016) may have brought zircons from such a far source area into the lower Danube's current basin. There is no obvious resolution to the question as to whether a previously continuous Paratethys could have transported laterally a significant amount of material from west of the Romanian Carpathians; there are no sedimentological data from the Dacian basin to support or refute that. However, a future study, perhaps a DZ study of the sedimentary archive of the Dacian basin, could resolve this question. This and the other puzzling complexities found in our data illustrate how the DZ record can be complicated by second or third order sedimentary processes that go beyond a simple (and direct) source to sink relationship in a fluvial system.

6. CONCLUSIONS

A detrital zircon U-Pb study of modern sands from the lower Danube and the most important four tributaries originating in the Carpathian Mountains documents the main magmatic events that led to the continental crustal formation of the nearby Carpathians. The main conclusions of this study are:

1- The great majority of basement was formed in latest Proterozoic – Ordovician island arcs, a finding that is consistent with limited previous studies performed on the basement itself;

2- A prominent Carboniferous (350-320 Ma, Variscan) magmatic peak in the detrital record has no known source in the nearby Carpathians, either because it was overlooked by previous basement studies or implying that lateral transport from outside of the source area (and subsequent recycling) has taken place in the recent geologic past. Some Variscan intrusions do exist in the Carpathians but according to the current geochronologic knowledge, are small volume plutons, and cannot account for such a large regional DZ peak. We cannot distinguish between these two explanations at this point due to limited existing data.

3- A small proportion of unexplained igneous Eocene ages exist along the Danube and tributaries; their closest exposed plausible sources are in the Rhodope Mountains well to the South without a clear sedimentary pathway from source to sink in the modern configuration of the river drainages.

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